Optimisation of ^{41}Ar environmental monitoring to provide a validation of radiological assessment models

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Abstract. The aerial discharges of particulate matter, from Sellafield, have declined by more than three orders of magnitude, since their peak in the mid 1970s, as a result of back-fitting abatement plant to the older installations. The decline in the level of particulates has led to the volatile and inert nuclides being the main contributors to the critical group dose. Of these ^{41}Ar, from the Calder Hall reactors, dominates with an assessed contribution of 42 μSv a^{-1} out of a total critical group dose of 65 μSv a^{-1} from both power generation and reprocessing discharges during 1999. This predominance has led to a requirement for field validation of the assessed dose. A programme of field measurements, using a sodium iodide spectrometer, calibrated to measure the external dose rate, has been carried out to determine the ^{41}Ar gamma flux at the critical group location. Comprehensive meteorological measurements were correlated with ^{41}Ar emission rates and gamma flux measurements. The resulting database of measurements has provided an extensive validation "toolkit" with which to determine the accuracy of the dose predictions from the radiological assessment models used by both industry and the UK regulators. This paper presents the results of the dose rate measurements of ^{41}Ar, together with a comparison of both long- and short-term dose rate predictions from a number of radiological assessment models. The relative performance of the models has been tested in a variety of meteorological stability conditions and discharge scenarios.

1. INTRODUCTION

Argon-41 (^{41}Ar) is present in aerial emissions from the Calder Hall reactors on the Sellafield site during routine operation. ^{41}Ar is a short-lived ($t_{1/2}$ = 1.83 hours) beta/gamma emitting radionuclide, produced by neutron activation of ^{40}Ar in the Calder Hall reactor cooling air. The discharges of ^{41}Ar are significant with 2,600 TBq released in 1999 [1]. Recent dose predictions using BNFL's Environmental Assessment Software (EAS) suggest that immersion in ^{41}Ar can account for up to 65% of the total adult terrestrial critical group dose from discharges [1]. Predicted doses resulting from exposure to ^{41}Ar are generally based on conservative modelled estimates, since measurement of ^{41}Ar in the field has been a difficult and time-consuming process. However, with new monitoring equipment designed to measure ^{41}Ar at low-level environmental concentrations, a programme of monitoring is now practicable.

This paper presents the results of a continuous monitoring programme to measure ^{41}Ar dose rates at the critical group location under a variety of meteorological and discharge conditions. Field measurements were compared against both long- and short-term model predictions from a number of radiological assessment models and the accuracy with which models predict the external dose was assessed.

2. MONITORING CAMPAIGN

2.1 The monitoring equipment

^{41}Ar external dose rates were measured using an Exploranium GR-320 γ-ray spectrometer located adjacent to the residence of the Calder Hall reactor critical group, which is within 1 km of the reactor stacks.

The GR-320 is configured to measure directly air kerma rates in nGy h^{-1} and was supplied pre-calibrated using US National Institute of Standards and Technology reference sources of ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{137}Cs, ^{54}Mn, ^{60}Co and ^{88}Y. The calibration procedure was based on that described by Grasty

[2] whereby an infinite cloud of radiation was simulated from measurements with radioactive sources placed on the surface of a sphere. Due to the axial symmetry of the cylindrical detector it was sufficient to measure the sources in a plane passing through the axis of the detector. Having determined the air kerma rate (Gy s^{-1}) at the centre of the crystal, for each calibration photopeak, the ratio of the air kerma rate to the counts per second under the photopeak was measured for each source/detector orientation along the surface of the sphere. A geometric weighting factor was applied to the ratio, calculated for each source/detector orientation, to give its contribution to the overall semi-infinite calibration factor.

During routine measurements in the field, the detector system used an automatic spectrum stabilisation facility set on the natural gamma ray peak at 2615 keV from ^{208}Tl in the thorium decay chain. The system is capable of resolving absorbed dose rates from ^{41}Ar of as little as 0.5 nGy h^{-1} in a sampling period of 15 minutes.

The procedure to remove the natural background variations from the ^{41}Ar photopeak was a modification of the standard stripping procedure used by field geologists to determine the ground concentrations of potassium, uranium and thorium. Counts in the uranium photopeak are used to remove increases in the ^{41}Ar peak background due to radon decay products that are brought down to the ground by rain or snow. Similarly the potassium photopeak is used to remove variations in the ^{41}Ar peak background due to fluctuations in soil moisture.

2.2 Meteorological data

Meteorological data was measured over the same period at the Sellafield meteorological compound, which is situated close to the Sellafield perimeter on the opposite side of the site to the Calder Hall reactor critical group. A 48 metre meteorological mast is equipped with anemometers, wind vanes and thermistors located at varying heights along the length of the mast. As well as providing wind speed and wind direction data for use in the models, temperature and wind speed profiles were used to calculate the turbulence characteristics of the atmospheric boundary layer as additional model input.

2.3 Stack emissions

Data on the magnitude of emissions of ^{41}Ar from the four Calder Hall reactors were obtained directly from the reactor physicist. Although not measured directly, the generation of ^{41}Ar from Calder Hall was reliably calculated from the known neutron flux and thus emission rates were estimated as a function of reactor power. The power output of the Calder reactors does not vary significantly throughout the day or night, therefore this method was considered to provide a reliable estimate of the ^{41}Ar discharge rates.

3. MODELLED DOSE RATES

The resulting database of ^{41}Ar field measurements has provided an extensive validation "toolkit" with which to determine the accuracy of the dose predictions from the assessment tools used by both industry and the UK regulators. In this study a number of atmospheric dispersion models and dose assessment tools were investigated under various meteorological conditions and for both short- and long-term discharge scenarios.

3.1 Short-term assessments

The accuracy of model predictions using EAS and UK-ADMS [3,4] were tested against field measurements. The main aim of this study was to determine the impact that different atmospheric dispersion models would have on the predicted air concentrations of ^{41}Ar, whilst the methodology used to estimate the dose was identical in both cases.

The atmospheric dispersion model contained within EAS is based on that described by Clarke [5] and widely used by the UK nuclear industry. It is a Gaussian plume model, where the horizontal and vertical concentrations are Gaussian in profile and the effects of atmospheric turbulence are included through the use of empirically derived stability classes.

UK-ADMS also describes atmospheric dispersion in terms of the standard deviations in the horizontal and vertical concentration distributions but the model uses a continuous scaling length, the Monin-Obukhov length, to derive atmospheric turbulence. In addition, a separate solution method, using a skewed concentration distribution, is applied to convective conditions, which are known to be significantly non-Gaussian. UK-ADMS is widely used in the UK by both regulators and consultants.

Although the UK-ADMS model has the functionality to calculate the gamma dose from immersion in a radioactive cloud, for this study the predicted air concentrations from UK-ADMS and EAS were used as input data to a semi-infinite cloud model. In this type of cloud model it is assumed that air concentration is uniform over the volume from which gamma photons can reach the point where dose is delivered and that the cloud is in radiative equilibrium.

The stack heights used in both models were derived from wind tunnel studies and are effective stack heights, which takes into account both the buoyancy of the plume and the downwash associated with adjacent buildings. The effective stack height used in EAS was 25 m, whilst for UK-ADMS the effective stack height was 20 m. The physical height of the stacks on the Calder Hall reactors from which ^{41}Ar is emitted is 50 m.

3.2 Annual assessments

In this case annual predictions of external dose were performed using EAS and PC-CREAM [6,7], a radiological assessment tool that is used widely by regulators to assess the impact from routine (annual) discharges. Unlike EAS, PC-CREAM uses a finite cloud model to calculate external dose. This method assumes that the distribution of activity in the plume is sufficiently non-uniform as to invalidate the semi-finite model approach. Consequently in the finite model the plume is divided into a series of volume sources and the effective photon flux from all the volume elements, which includes both the scattered and unscattered components, is obtained by integrating over space. The effective photon flux is converted to absorbed dose in air and then effective dose by use of the appropriate conversion factors [7].

4. RESULTS

4.1 Measured external dose rates

Figure 1 shows the ^{41}Ar dose rates (nSv h^{-1}) measured at the Calder Hall reactor critical group location for the period 14th September 2000 until 18th February 2001. Monitoring was stopped prematurely due to the outbreak of Foot and Mouth disease in the United Kingdom, which prevented access to the site. However, during this period over 8,000 gamma-ray spectra were collected over a period of 3,634 hours, resulting in a measured air kerma rate of 5.5 nGy h^{-1}. The absorbed dose in air was converted to an effective dose of 4.0 nSv h^{-1} using a conversion factor (Sv Gy^{-1}) that relates absorbed dose in air as a function of the initial photon energy [7].

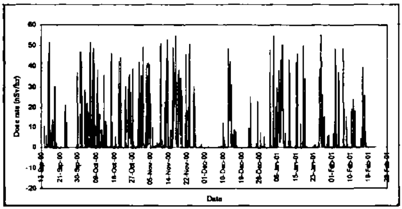

Figure 1: Graph of measured ^{41}Ar dose rates (nSv h^{-1}) at the critical group location for the period 14th September 2000 - 18th February 2001

The unshielded dose measured during the monitoring period was thus 15 µSv, which if extrapolated for the year would produce an unshielded annual adult dose of 35 µSv. It is assumed that the adult member of the critical group spends 66% of their time indoors, and the building will offer some protection from irradiation. Consequently a building-shielding factor of 0.2 [7] can be applied which will result in an annual dose of 17 µSv a^{-1}, arising from an extrapolated discharge of approximately 2,760 TBq a^{-1} of ^{41}Ar.

4.2 Model comparison – annual assessments

The annual doses predicted by EAS and PC-CREAM, based on a discharge of 2,760 TBq a^{-1} of ^{41}Ar, were 45 µSv and 42 µSv respectively. Comparing these predictions with the extrapolated measured dose shows that EAS and PC-CREAM over-predict the annual dose by factors of 2.6 and 2.5 respectively. This over-prediction is partly due to the fact that estimates of annual dose are derived by model configurations that use generalised input data and parameter values.

4.3 Model comparison – short-term assessments

Several statistical tests were performed to assess how well the models EAS and UK-ADMS predict the hourly ^{41}Ar dose rates. The R^2 statistic describes the scatter of the modelled doses about a linear regression line. The Normalised Mean Square Error (NMSE) test was used to quantify the scatter in modelled values about the true measured data (rather than about a regression line) and therefore provides information on the precision of the results. It should be noted that the NMSE is a function of the range of measured values being compared and so can only be used to compare model predictions for the same data sets. An NMSE value of 0.0 represents perfect precision of the modelled and measured dose rate.

The Mean Bias (MB) was calculated as the ratio of the mean of the measured dose rates to the mean of the predicted. The mean bias therefore represents the over-prediction or under-prediction of the model, as it compares the mean of all the valid measured data to the mean of all the valid modelled results. A value of 1 for the MB represents a perfect match between the modelled and measured doses.

To assess the accuracy of the predictions, the fraction of modelled values within factors of 2, 5 and 10 of the measured values were calculated and are reported as the F2, F5 and F10 statistics respectively.

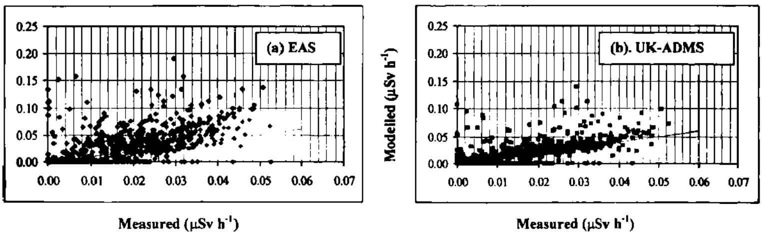

Figure 2a, b: Plots of measured ^{41}Ar dose rates (µSv h^{-1}) versus predicted using EAS and UK-ADMS

Figures 2a and 2b are graphs of measured dose rate versus those predicted using EAS and UK-ADMS respectively. In this comparison all the available measurements were used. The straight line on the scatter plots represents a perfect fit between the measured and modelled data. Figure 2 illustrates that in general both models over-predict dose rate, but that UK-ADMS performs better than EAS. This is reflected in table 1, which lists the statistical comparisons between the measured and predicted values and shows that UK-ADMS over-predicts the dose by 17% (MB=1.17), whilst EAS over-predicts by 60%

(MB=1.60). In addition, the F2, F5 and F10 statistics show that UK-ADMS out-performs EAS with 57% of the predictions from UK-ADMS being within a factor of 2 of the measurements compared to only 35% from EAS.

The database of measurements and predictions can be interrogated further to investigate those parameters and conditions that influence the calculated dose rates. For example, during neutral stability conditions, when dispersion is dominated by mechanical mixing, both models show a marked improvement in performance with less scatter in the predictions. Data in table 1 shows that in the case of EAS, 54% of the predictions are within a factor of 2, whilst 90% are within a factor of 10 and the NMSE is now less than 1.0. For UK-ADMS, 83% of the model predictions are within a factor of 2 of the measurements, whilst almost 100% of the predictions are within a factor of 5 and the NMSE is now 0.27.

Table 1: Statistical analysis of the short-term ^{41}Ar dose rates predicted by EAS and UK-ADMS

Model Run	No. of spectra	Mean (μSv h^{-1})			MB		R^2	
		Measured	EAS	ADMS	EAS	ADMS	EAS	ADMS
All data	3383	4.36E-03	7.14E-03	5.20E-03	1.60	1.17	0.41	0.47
Cat. A-C	237	4.49E-03	1.28E-02	5.99E-03	2.85	1.33	0.10	0.06
Cat. D	1889	5.05E-03	8.49E-03	6.14E-03	1.68	1.21	0.50	0.59
Cat. E-G	1103	3.23E-03	3.32E-03	3.30E-03	1.03	1.02	0.36	0.42
> 2m/s	2783	4.60E-03	7.64E-03	5.51E-03	1.66	1.20	0.50	0.54
Model Run	**F2**		**F5**		**F10**		**NMSE**	
	EAS	**ADMS**	**EAS**	**ADMS**	**EAS**	**ADMS**	**EAS**	**ADMS**
All data	0.35	0.57	0.60	0.70	0.65	0.73	1.64	0.74
Cat. A-C	0.19	0.41	0.53	0.68	0.69	0.75	3.68	1.56
Cat. D	0.54	0.83	0.84	0.96	0.90	0.98	0.91	0.27
Cat. E-G	0.09	0.18	0.20	0.28	0.23	0.31	4.03	2.59
> 2m/s	0.42	0.67	0.70	0.82	0.75	0.84	1.24	0.49

The models also perform relatively well if predictions made during low wind speeds (i.e. <2ms^{-1}) are excluded from the analysis since Gaussian plume models are known to perform poorly in these conditions.

In unstable conditions (Cat. A-C) i.e. when dispersion is dominated by convection, the models severely over-predict and even in the case of UK-ADMS, which includes algorithms specific for convective dispersion, only 41% of the predictions are within a factor of 2 of the measurements.

However, during stable conditions (Cat. E-G) the performance of the models deteriorates to a point where only 9% of EAS and 18% of UK-ADMS predictions are within a factor of 2 of the observations.

5. CONCLUSIONS AND SUMMARY

The total measured dose recorded at the Calder Hall reactor critical group was 15 μSv which was extrapolated to produce an annual adult shielded dose of 17 μSv a^{-1} from an estimated discharge of approximately 2,760 TBq a^{-1} of ^{41}Ar. Comparing the measured dose with that predicted using EAS (i.e. 45 μSv a^{-1}) and PC-CREAM (i.e. 42 μSv a^{-1}) it is found that the calculated doses are over-predicted by factors of 2.6 and 2.5 respectively. If the extrapolated dose is a true representation of the ^{41}Ar dose, then it approaches a threshold (20 μSv a^{-1}), at which UK regulators will not seek further reduction in public exposures, provided that 'Best Practicable Means' are being applied to safeguard the public [8].

Comparison of observations with short-term dose predictions using EAS and UK-ADMS indicates that the calculated dose rate is once again over-predicted, though to a lesser extent than for annual assessments. In the case of EAS, which uses a simple Gaussian plume model common to many radiological assessment tools, the dose rate can be overestimated by as much as 60%. Under neutral stability conditions both EAS and UK-ADMS performed well, however, the accuracy of dose rate predictions deteriorates during unstable and stable meteorological conditions or at low wind speeds.

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